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(54) Title: POLYMERIC THICKENERS FOR OIL-CONTAINING COMPOSITIONS

(57) Abstract: Thickeners for oil-containing compositions are side chain crystalline polymers, which (a) are uniformly dispersed in the oil as a crystallized solid, (b) are soluble in the oil at temperatures above T<sub>p</sub>, and (c) are substantially free of carboxyl groups, carboxyl groups in the form of salts, sulfonic acid groups, and sulfonic acid groups in the form of salts.

## POLYMERIC THICKENERS FOR OIL-CONTAINING COMPOSITIONS

5 This invention relates to polymeric thickeners for oil-containing compositions.

U.S. Patent Nos. 4,057,622, 4,057,623, 4,057,624, 5,318,995, 5,519,063 and  
5,736,125 disclose the possibility of thickening oil-containing compositions with  
polymers containing lipophilic groups (e.g. in units derived from n-alkyl acrylates) and  
10 carboxylic or sulfonic acid groups, or salts thereof (e.g. in units derived from acrylic or  
methacrylic acid).

We have discovered, in accordance with present invention, that oils can be  
thickened by side chain crystalline (SCC) polymers which do not contain carboxyl or  
15 sulfonic acid groups, or salts thereof. Such SCC polymers must, however, be dispersed  
in the oil as a crystallized solid, in order to produce effective thickening. The SCC  
polymer is preferably dispersed in the oil by

- (i) dissolving the SCC polymer in the oil at a temperature above the  
crystalline melting point of the polymer, and
- 20 (ii) cooling the solution to crystallize the polymer in the oil.

We believe (though the invention is not dependent upon the correctness of our belief)  
that in step (ii) the SCC polymer crystallizes into a network in which the polymer  
crystallites are connected to one another by semi-soluble chains.

25 In one preferred aspect, this invention provides a thickened oil composition which  
is at a temperature  $T_s$  and which comprises

- (1) an oil, and
- (2) uniformly dispersed in the oil as a crystallized solid, a side chain  
crystalline (SCC) polymer having a crystalline melting point,  $T_p$ , which is  
30 greater than  $T_s$ ;

the composition being substantially free of polymers containing carboxyl groups,  
carboxyl groups in the form of salts, sulfonic acid groups, and sulfonic acid groups in the  
form of salts. The composition must be maintained at a temperature below  $T_p$ , since the

polymer will cease to have a thickening effect if it redissolves in the oil. The composition can be free of water, or can be a water-in-oil emulsion.

One of the advantages which results from the use of these SCC polymers as thickening agents is that they reduce or remove the need to use surface active agents in water-in-oil emulsions. This is particularly useful in personal care products, since it is conventional for such products to contain surfactants which can cause an adverse reaction when they contact human skin.

## 10 Definitions and Abbreviations

In this specification, parts and percentages are by weight. Temperatures are in °C. The onset-of-melting temperature,  $T_o$ , the peak melting temperature,  $T_p$ , and the heat of fusion, J/g, are determined using a differential scanning calorimeter (DSC) at a rate of temperature change of 10 °C./min, for example from -10 to 150 °C., and on the second heat cycle).  $T_p$  is the temperature at the peak of the DSC curve, and  $T_o$  is the temperature at the intersection of the baseline of the DSC peak and the onset line, the onset line being defined as the tangent to the steepest part of the DSC curve below  $T_p$ . The abbreviations  $M_n$  and  $M_w$  are used to denote number average and weight average molecular weight in daltons, respectively, measured in tetrahydrofuran using size exclusion chromatography, configured with a Wyatt laser light scattering detector. Bulk viscosities given in the Examples for the polymeric thickeners are in centipoise and were measured using a Brookfield LVT viscometer with an electrically thermostatted Thermosel heater, controlled for example to 95 °C., and small sample adapter using spindles 4 and 7.

The procedures used in the Examples to compare the effectiveness of the polymeric thickeners were as follows. In Examples 1-8, the thickener, 5 parts, was dissolved in hydrogenated polyisobutylene (HPIB, a light oil), 95 parts, with stirring at 120 °C. The resulting solution was placed in an incubator at 20 °C. for 16 hours. The viscosity of the cooled product in centipoise was determined using a Brookfield DV-1+ digital viscometer with CP-51 spindle using a sample adapter which was thermostatically controlled, for example, to 25 °C. The viscosities were measured after

four minutes at a speed of 2.5 rpm, i.e. after 10 revolutions. In Examples 9-17, the oil (as identified in Table 2), 14 parts, was heated to 80 °C., and the thickener, 0.75 part, was dissolved therein. Water, 35 parts, containing  $\text{MgSO}_4 \cdot \text{H}_2\text{O}$ , 0.25 part, was heated to 80 °C. The oil and the water, both at 80 °C, were mixed together, and then cooled to 25 °C. with continued stirring. The product, a milky white water-in-oil emulsion, was left overnight, and its viscosity at 25 °C. was then measured using a Brookfield cone and plate viscometer. The viscosity was measured after 0.5, 1, 2 and 4 minutes, to assess the effect of shear on the emulsion.

The abbreviation CxA is used to denote an n-alkyl acrylate in which the n-alkyl group contains x carbon atoms, the abbreviation Cx alkyl is used to denote an n-alkyl group which contains x carbon atoms, and the abbreviation CxM is used to denote an n-alkyl methacrylate in which the n-alkyl group contains x carbon atoms. Other abbreviations are given elsewhere in the specification.

#### The Polymeric Thickeners

The SCC polymers used as thickeners in the present invention can be homopolymers, or copolymers of two or more comonomers, including random copolymers, graft copolymers and block copolymers (including thermoplastic elastomers). Two or more SCC polymers can be used together. The number average molecular weight of the SCC polymer is generally from 10,000 to 1,500,000, preferably 12,000 to 1,000,000. The molecular weight of an SCC polymer is relatively unimportant to its  $T_p$ , but is generally an important factor in determining the  $T_p$  of other polymers.

The SCC polymer preferably melts over a relatively small temperature range. The closer  $T_p$  is to room temperature, the more rapid the transition should preferably be. The SCC polymer preferably has an onset of melting temperature,  $T_o$ , such that  $T_p - T_o$  is less than  $T_p^{0.7}$ , generally less than  $T_p^{0.6}$ , particularly less than 10 °C., especially less than 6 °C.,  $T_o$  and  $T_p$  being in °C. The crystallinity of the SCC polymer is preferably such that its heat of fusion is at least 20 J/g, particularly at least 40 J/g.

The SCC polymers used in the present invention are in themselves well known. Publications describing SCC polymers include U.S. Patent Nos. 4,830,855, 5,120,349, 5,156,911, 5,387,450, 5,412,035, 5,665,822, 5,783,302, 5,752,926, 5,807,291, 5,469,867, and 5,826,584; J. Poly. Sci. 60, 19 (1962), J. Poly. Sci. (Polymer Chemistry) 7, 3053  
5 (1969), 9, 1835, 3349, 3351, 3367, 10, 1657, 3347, 18, 2197, 19, 1871, J. Poly. Sci., Poly-Physics Ed 18 2197 (1980), J. Poly. Sci. Macromol. Rev, 8, 117 (1974), Macromolecules 12, 94 (1979), 13, 12, 15, 18, 2141, 19, 611, JACS 75, 3326 (1953), 76, 6280, Polymer J 17, 991 (1985); and Poly. Sci USSR 21, 241 (1979).

10 The SCC polymer may for example be derived from one or more acrylic, methacrylic, olefinic, epoxy, vinyl, ester-containing, amide-containing or ether-containing monomers. Preferred SCC polymers comprise repeating units in which the side chains comprise linear polymethylene radicals containing 10 to 50, e.g. 16-50, especially 16 to 22, carbon atoms, or linear perfluorinated or substantially perfluorinated polymethylene  
15 radicals containing 6 to 50 carbon atoms. Polymers containing such units can be prepared by polymerizing a monomer component comprising one or more corresponding linear aliphatic acrylates or methacrylates; or equivalent monomers such as acrylamides or methacrylamides. A number of such monomers are available commercially, either as individual monomers or as mixtures of identified monomers, for  
20 example C12A, C14A, C16A, C18A, C22A, a mixture of C18A, C20A and C22A, a mixture of C26A to C40A, fluorinated C8A (AE800 from American Hoechst) and a mixture of fluorinated C8A, C10A and C12A (AE812 from American Hoechst). The polymers may also contain units derived from one or more other comonomers, for example straight or branched chain alkyl acrylates or methacrylates in which the alkyl  
25 group contains less than 12 carbon atoms, and monomers containing desired functional groups, for example the monomers listed below; and equivalent monomers such as acrylamides and methacrylamides. In the list below, the term (meth) acrylate means that the compound may be either an acrylate or a methacrylate.

(a) Nitrogen-containing monomers, for example N,N-dialkyl amino (in particular,  
30 dimethylamino) (meth)acrylates; ammonium salt-containing (meth) acrylates, for example 2-trimethylammonium methylemethacrylate chloride,

methacrylamidopropyl trimethylammonium chloride, N,N- (diethyl or dimethyl)aminoethyl(meth)acrylate methosulfate; N-vinylpyrrolidinone; imides like the ring-closed reaction products of maleic or itaconic anhydride with primary amines; 2-methacryloxy-N-ethylmorpholine; n or t-butylacrylamide; (meth)acrylamide; dimethylaminopropyl methacrylamide; 2-t-butylaminoethyl methacrylate; (meth)acrylonitrile; t-butylaminoethyl (meth)acrylate; acryloylmorpholine; N-(2-hydroxyethyl)acetamide and 1-piperidinoethyl (meth)acrylate.

(b) Oxygen-containing monomers (other than carboxyl- and sulfonic acid-containing monomers and salts thereof), for example hydroxyalkyl (in particular, hydroxyethyl, hydroxypropyl, and hydroxybutyl) (meth)acrylates; tetrahydrofurfuryl (meth)acrylate; glycidyl methacrylate; alkoxyalkyl (meth)acrylate, e.g. methoxyethyl (meth)acrylate; 1-acryloxy-2-hydroxy-3-phenoxypropane; methylol methacrylate; ethoxyethyl (meth)acrylate; 2-(2-ethoxyethoxy)ethylacrylate; acetoacetoxyethyl (meth)acrylate; phenoxyethyl (meth)acrylate and (meth)acrolein.

(c) Fluorine-containing monomers, for example trifluoroethyl (meth)acrylate, heptadecafluorodecyl (meth)acrylate, octafluoropentyl (meth)acrylate, eicosafuoroundecyl (meth)acrylate, hexadecafluorononyl (meth)acrylate, and tetrahydroperfluorodecyl (meth)acrylate.

(d) Silicon-containing, e.g. silyl, monomers, for example trimethylsiloxy ethyl(meth)acrylate, 3-acryloxypropyl trimethoxysilane, and 3-acryloxypropyl tris(trimethylsiloxy)silane.

When the SCC polymer is a graft or block copolymer, it can be prepared by copolymerizing a vinyl type macromonomer with other monomers, or by making an SCC polymer, and then reacting the functionalized polymer with the second block material, for example a urethane block, or an epoxy block, a polyether block, e.g. a polyethyleneoxide, polypropyleneoxide or polytetramethyleneoxide block, a polysiloxane block, or a poly(alkyl or alkoxy)silane block.

The SCC polymer should contain sufficient long chain groups that it will dissolve in the oil at a temperature above  $T_p$ . When the SCC polymer is used to thicken an oil or mixture of oils which is free from water, it generally contains at least 50 %, preferably at least 60%, particularly at least 70%, especially at least 80%, of units derived from a monomer containing 10 to 50 carbon atoms, and can contain up to 100% of such units. When the SCC polymer is used to thicken a water-in-oil emulsion, it may contain at least 5%, preferably at least 10%, of units derived from a monomer containing a hydrophilic group, and may contain higher amounts, e.g. up to 25%, provided that the SCC polymer will dissolve in the oil.

The molecular weight of the thickening polymer should be sufficiently high that the polymer, after it has been dissolved in the oil, will crystallize in the oil when the heated mixture is cooled, for example to a temperature 10-20 °C. below  $T_p$ , thus producing an opaque mixture.

The  $T_p$  of the thickening polymer is preferably 10-40 °C. above, particularly 10-30 °C. above, especially about 20 °C. above, the temperature at which the composition is to be used. It appears that the oil plasticizes the thickening polymer, so that its melting point in the composition is for example 5-10 °C. lower than  $T_p$ . It is therefore important that  $T_p$  is sufficiently above the temperature of use to ensure that the thickening polymer does not melt during use. Thus for compositions to be used at 20-25 °C. the thickening polymer preferably has a  $T_p$  of above 40 °C., preferably 40-50 °C. On the other hand, if the  $T_p$  of the thickening polymer is too far above the temperature of use, this can result in excessive crystallization and then precipitation of the polymer, thus reducing the thickening effect. It is preferred, therefore, that  $T_p$  is not more than 30 °C., preferably not more than 20 °C., above the temperature of use. Depending on the expected temperature of use,  $T_p$  may be from 0-150 °C. generally 10-100 °C., for example 40-80 °C.

The amount of the polymeric thickener preferably used varies with the application. It is usually unnecessary for the amount of the thickener to be more than

10% of the total composition, and smaller amounts such as 2 to 7% in compositions which are free of water, and 0.5 to 5% in water-in-oil emulsions, are often effective.

### Oils

5

The new polymeric thickeners are effective with a broad range of oils. Suitable oils are disclosed, for example, at column 3, line 37, to column 4, line 4, of U.S. Patent No. 5,736,125. For thickening silicone oils, it is preferred to use an SCC polymer containing units derived from a monomer containing silicon, for example a block  
10 copolymer containing SCC blocks and polysiloxane blocks. SCC/polysiloxane polymers of this type are described for example in WO 93/07194 and WO 00/04787.

### Water-in-oil Emulsions

15 Water-in-oil emulsions are preferably prepared by mixing together (1) a hot solution of the thickener in the oil and (2) the aqueous phase, the aqueous phase being at a temperature similar to the oil solution (e.g. not more than 10 °C. different); and then cooling the mixture while stirring. The ratio of the aqueous phase to the oil phase can be for example 0.5:1 to 9:1.

20

### Compositions

The invention is particularly useful for personal care compositions, for example cosmetics, toiletries, and cleansers, including but not limited to lipsticks, deodorant  
25 sticks, nail varnishes, creams, gels and oils, including sun creams, protective hand creams, night renewal creams, body milks and lotions, light facial cream, protective day cream, liquid moisturizing emulsions, oil-in-water creams and water-in-oil creams, as well as thickened oil products with or without water and products designed to assist in removing other cosmetic, makeup or personal care products. The invention is also  
30 useful in other contexts, for example in paints, film-forming compositions, inks, and compositions carrying active ingredients such as UV absorbers, fragrances, biocides,



antimicrobial agents, germicides, antioxidants, preservatives, disinfectants, enzymes, nutrients, minerals, and drugs (including pharmaceuticals which are active physiologically or pharmacologically, either topically or systemically). Compositions containing a thickener containing an ammonium salt are likely to be useful in certain types of hair care compositions.

The invention is illustrated by the following Examples.

### Examples

10

Polymers and copolymers were made using the ingredients and amounts thereof shown in the Table below, using the following generalized method. To a resin kettle equipped with overhead stirrer and condenser was added 20% of the monomers and chain transfer agents. The mixture in the resin kettle was heated to 110 °C., and oxygen was removed from the system through nitrogen purge for about 30 min followed by addition of 20% of the starting initiator charge. After allowing sufficient time for any initial exotherm to abate, the remaining monomers, chain transfer agents and starting initiator were pumped into the reaction vessel over 60-90 min. The polymer mixture was allowed to continue reacting for 60 min followed by addition of the chase initiator and reaction for 60 min. The mixture was put under reduced pressure for 60 min to removal volatile residuals. The resulting polymers were generally yellow to white solids.

The molecular weight,  $T_p$ , and viscosity of each sample were measured. The effectiveness of the polymers as thickeners was measured as described above, and the results are shown in Tables 1 and 2 below.

25

The following abbreviations are used in the Tables. ME = mercaptoethanol; MA = methacrylic acid; NVP = N-vinylpyrrolidinone; DMAEA = N,N-dimethylaminoethyl acrylate; HEA = 2-hydroxyethyl acrylate; TAPO = t-amylperoxy 2-ethylhexanoate sold by Witco as Esperox 570P, 75% active in liquid; TBPB = t-butylperoxybenzoate sold by Witco as Esperox 10; Estol is propylene glycol dicaprylate/caprate sold by Uniqema under the tradename Estol 1526; Min'l is mineral oil; and opq = opaque in appearance.

30

TABLE 1

Example	1	2*	3*	4*	5	6	7	8
C16A				95				
C18A	80		95		100	85	85	85
C22A		95						
HEA	20							
NVP						15	15	
DMAEA								15
MA		5	5	5				
ME	0.34	0.17	0.17	0.17	0.17	0.34	0.34	0.1
TAPO	1.73	1.73	1.73	1.73	1.73	1.73	1.73	1.73
TAPB	0.67	0.67				0.67	0.67	
TBPB			0.5	0.5	0.5			0.5
T <sub>p</sub> °C.	48	67	47	39	50	48	46	45
J/g	56	99	57	64	73	29	61	60
M <sub>w</sub>	236K		427K	1,000K	950K			
M <sub>n</sub>	52K		240K	520K	230K			
Bulk viscosity	4,000	2,500	19,000	24,000	2,000	3,950	4,000	350
Visc in HPIB	12,600 opq	164 opq	2600 opq	<50 clear	5400 opq	5100 opq	6200 opq	6000 opq

\* Examples 2, 3 and 4 are comparative examples in which the thickener contained units derived from methacrylic acid. In Example 2, the polymer had a T<sub>p</sub> above the preferred range of 40-50 °C., which resulted in excessive crystallinity and poor thickening. In Example 4, the polymer had a T<sub>p</sub> below the preferred range, and was ineffective as a thickener because it did not crystallize on cooling. In Example 3, the T<sub>p</sub> of the polymer was in the preferred range, but the polymer was less effective than the polymers of the invention used in Examples 1 and 5-8.

TABLE 2

Example	9	10	11	12	13	14	15	16	17
C18A	80	80	80	85	85	85	100	100	100
HEA	20	20	20						
NVP				15	15	15			
ME	0.34	0.34	0.34	0.34	0.34	0.34	0.17	0.17	0.17
TAPO	1.73	1.73	1.73	1.73	1.73	1.73	1.73	1.73	1.73
TAPB	0.67	0.67	0.67	0.67	0.67	0.67			
TBPB							0.5	0.5	0.5
Tp °C.	48	48	48	48	48	48	50	50	50
J/g	56	56	56	29	29	29	73	73	73
Mw	236K	236K.	236 K				950 K.	950 K.	950 K.
Mn	52K	52 K.	52 K.				230 K.	230 K.	230 K.
Bulk viscosity	4,000	4,000	4,000	3,950	3,950	3,950	2,000	2,000	2,000
Oil	HPIB	Min'l	Estol	HPIB	Min'l	Estol	HPIB	Min'l	Estol
Viscosity after									
0.5 min	35K	37K	45K	36K	42K	48K	29K	**	**
1.0 min	35K	38K	40K	35K	44K	50K	28K		
2.0 min	41K	38K	34K	23K	38K	40K	25K		
4.0 min	46K	40K	34K	18K	22K		28K		

\*\* separated.

- 5 The invention has been described by reference to the use of SCC polymers. However, other polymers with the same crystallinity characteristics give similar results, and their use as thickeners for oils forms part of the present invention. Such other polymers include for example polymers in which the crystallinity results exclusively or predominantly from the polymer backbone, e.g. polymers of  $\alpha$ -olefins containing 2 to 12, preferably 2 to 8, carbon atoms, e.g. polymers of monomers having the formula  $\text{CH}_2 = \text{CHR}$ , where R is hydrogen, methyl, propyl, butyl, pentyl, 4-methylpentyl, hexyl or heptyl, as well as other polymers such as polyesters, polyamides, for example homopolymers and copolymers of caprolactone, and polyalkylene oxides, for example polytetrahydrofuran. The invention also includes the use as thickeners for oils of block and graft copolymers comprising (i) SCC polymeric moieties which contain carboxylic or sulfonic acid groups, or salts thereof, and (ii) other polymeric moieties, in particular polysiloxane and/or polyether moieties, which are not disclosed in the prior art.

## CLAIMS

1. A thickened oil composition which is at a temperature  $T_s$  and which comprises
- (1) an oil, and
  - (2) uniformly dispersed in the oil as a crystallized solid, a side chain crystalline (SCC) polymer having a crystalline melting point,  $T_p$ , which is greater than  $T_s$ ;
- the composition being substantially free of polymers containing carboxyl groups, carboxyl groups in the form of salts, sulfonic acid groups, and sulfonic acid groups in the form of salts.
2. A composition according to Claim 1, wherein the SCC polymer consists of repeating units in which the side chains comprise unsubstituted alkyl groups.
3. A composition according to Claim 1 or 2, wherein the SCC polymer is a homopolymer, a block copolymer or a graft copolymer.
4. A composition according to Claim 1, wherein the SCC polymer contains repeating units containing an oxygen-containing, nitrogen-containing, fluorine-containing or silicon-containing group.
5. A composition according to Claim 4, wherein the SCC polymer contains 10-30% of repeating units containing hydroxyl groups.
6. A composition according to any one of the preceding claims, wherein the SCC polymer is present in amount at least 3 % by weight and contains at least 70% by weight of repeating units containing a side chain comprising a linear polymethylene radical containing 16 to 50 carbon atoms or a linear perfluorinated polymethylene radical containing 6 to 50 carbon atoms.

7. A composition according to any one of the preceding claims, wherein the SCC polymer has a  $T_p$  above 40 °C. and a heat of fusion of at least 20 J/g.
8. A composition according to any one of the preceding claims, wherein
- 5 (a) the SCC polymer has a  $T_p$  of 40-50 °C.;
- (b) 70-100% of the repeating units in the SCC polymer are derived from at least one acrylate or methacrylate ester in which the ester group contains an unsubstituted alkyl group; and methacrylamide
- 10 (c) 0-25% of the repeating units in the SCC polymer are derived from at least one acrylate or methacrylate ester in which the ester group contains a hydroxyl-substituted alkyl group.
9. A method of making a composition according to any one of the preceding claims, the method comprising
- 15 (i) dissolving the SCC polymer in the oil at a temperature above  $T_p$ , and
- (ii) cooling the solution to crystallize the polymer in the oil.
10. A method according to Claim 9, wherein
- (1) the SCC polymer has an onset of melting temperature,  $T_o$ , such that
- 20 ( $T_p - T_o$ ) is less than  $T_p^{0.7}$ ,  $T_o$  and  $T_p$  being measured in °C.; and
- (2) the SCC polymer has at least one of the following characteristics
- (i) it contains an oxygen-containing, nitrogen-containing, fluorine-containing or silicon-containing group;
- (ii) it contains repeating units containing long chain alkyl
- 25 groups containing at least 26 carbon atoms;
- (iii) it is substantially free of functional groups;
- (iv) it is substantially free of ionizable groups;
- (v) it is a block copolymer or a graft copolymer; and
- (vi) it is a homopolymer.
- 30

# INTERNATIONAL SEARCH REPORT

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> IPC 7 A61K7/48		International Application No <b>PCT/US 00/40780</b>
According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) IPC 7 A61K C08L		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal, WPI Data, PAJ		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 5 736 125 A (N. MORAWSKY ET AL.) 7 April 1998 (1998-04-07) cited in the application claims	1-10
Y	WO 96 27641 A (LANDEC CORPORATION) 12 September 1996 (1996-09-12) page 3, line 7 - line 8; claims page 12, line 18 -page 15, line 19	1-10
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A	EP 0 803 513 A (KAO CORP) 29 October 1997 (1997-10-29) claims	1
<div style="display: flex; justify-content: space-between;"> <span><input type="checkbox"/> Further documents are listed in the continuation of box C.</span> <span><input checked="" type="checkbox"/> Patent family members are listed in annex.</span> </div>		
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Date of the actual completion of the international search <div style="text-align: center; font-weight: bold;">13 February 2001</div>		Date of mailing of the international search report <div style="text-align: center; font-weight: bold;">20/02/2001</div>
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax (+31-70) 340-3016		Authorized officer <div style="text-align: center; font-weight: bold;">Boeker, R</div>

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Information on patent family members

International Application No

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